Interactions between columnar solidification and segregation: a comparison of the predictions of the CAFE model with in-situ observations

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INTERACTIONS BETWEEN COLUMNAR SOLIDIFICATION AND SEGREGATION: A COMPARISON OF THE PREDICTIONS OF A CAFE MODEL WITH IN-SITU OBSERVATIONS

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Abstract

A two-dimensional (2D) cellular automaton (CA) - finite element (FE) model has been proposed to simulate the solidification of binary alloys. The time evolution of maps of temperature, composition and fraction of solid are simulated while accounting for the undercooling of the growing structure. The solidification experiment of a Gallium-5wt%Indium alloy developed by Yin and Koster [1-2] is shown to provide a good benchmark for comparison with predictions of the model. Observed and simulated time evolutions of the development of the grain structure and the macrosegregation can be directly compared, together with mesosegregation such as segregated channels. These channels differ from freckle-type segregation described in the literature. They form as a result of the dynamics of the columnar growth front and its interaction with the fluid and solute flows. Comparisons of the predictions of the CAFE model with a purely macroscopic FE model also reveal the influence of accounting for the growth undercooling in numerical modeling of solidification.

Introduction

Prediction of as-cast structure and segregation in ingot is part of the objectives of modeling tools. These features are linked to the quality of casting processes and soundness of cast parts. Several models have been developed over the years for the prediction of structure formation in casting [3-7]. Cellular Automaton (CA) - Finite Element (FE) models are particularly well suited to track the development of a columnar dendritic front in an undercooled liquid at the scale of the casting [5-7]. Although these models do not directly describe the complicated nature of the solid-liquid interface that defines the dendritic microstructure, the crystallographic orientation of the grains as well as the effect of the fluid flow can be accounted for to calculate the undercooling of the mushy zone growth front. Two- and three-dimensional CAFE models were successfully applied to predict features such as the columnar-to-equiaxed transition observed in Aluminum-Silicon alloys [5], the selection of a single grain and its crystallographic orientation due to the competition among columnar grains taking place while directionally solidifying a

superalloy into a pig-tail shape [6], as well as the effect of the fluid flow on the fibre texture selected during columnar growth [7]. Recently coupling with macrosegregation has been developed [8], thus providing with an advanced CAFE model to account for structure formation compared to purely macroscopic models developed so far, e.g. [9-11]. While both structure and segregation are predicted, comparisons with the experimental results concerning the structure were yet limited due to a lack of detailed data of experimental results [12]. In particular, comparison is mainly conducted with the as-cast state.

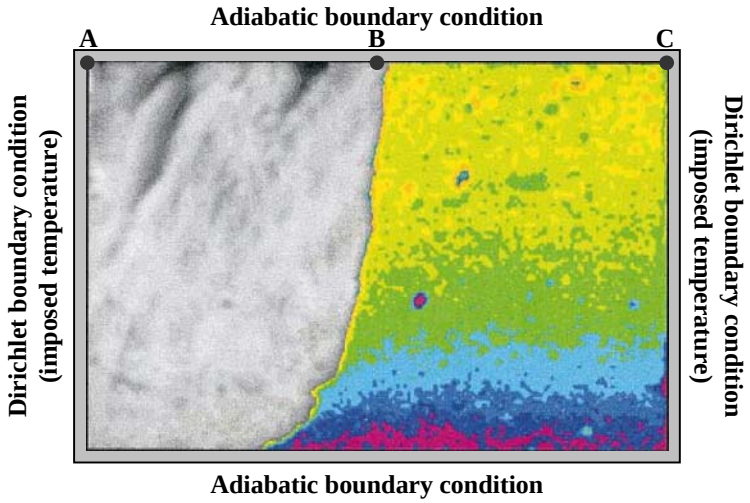

Figure 1. Formation of Indium stratification in the liquid (horizontal bands in the right-hand-side) and corresponding growth front morphology 165 minutes after beginning the cooling of a Gallium-5wt%Indium alloy as observed by Yin and Koster [1-2]. Dimension of the field of view: 0.048 m width × 0.033 m height. Dots labeled A-C: location of thermocouples.

This contribution presents a first comparison of the predictions of the CAFE model with *in-situ* observations of structure and segregation developing in a Gallium-5wt%Indium alloy as carried out by Yin and Koster [1-2]. A snapshot of the observation made by these authors is shown in Figure 1 at a given time during solidification. The experimental apparatus consists of a parallelepipedic mold with inner dimensions 0.050 m width × 0.035 m height × 0.002 m thick. Edges of the samples are hidden thus leaving a field of view with dimensions 0.048 m width × 0.033 m height. The main faces of the mold as well as the bottom surface are considered as adiabatic. Despite the presence of a reservoir at the top part of the mold, its geometry is designed to limit exchanges and an adiabatic condition can also be assumed. The temperature evolution of thermostats in contact with the left- and right-hand-side walls of the system is controlled. Overall, the cooling of the sample is thus essentially two-dimensional. In this paper is presented a comparison of CAFE predictions with the well documented experimental case "SOL-1" as named by Yin and Koster [1]. The recorded evolutions of the temperature at location of thermocouples A and C displayed in Figure 1 is imposed as Dirichlet boundary conditions. In-situ X-Ray radioscopic visualization of the ingot was performed during the solidification experiment. The images clearly reveal the progression of the structure through the cavity. A single grain is found to nucleate at a measured undercooling and grow from the left-hand-side wall at time 110 minutes, with clear anisotropic growth direction. An orientation between the main trunk direction of the crystal and the horizontal axis is measured and imposed for nucleation in the simulation. Further analysis of the X-Ray images is performed to deduce composition maps. Indium is found to be mostly distributed as horizontal fringes in the liquid

located in the right-hand-side of Figure 1, highly segregated at bottom and slightly segregated at top. This is explained by mushy zone segregation and solute-driven convection. The various grey levels in the mushy zone located on the left-hand-side of Figure 1 are also related to the average Indium composition. The darker the grey level is, the higher the composition.

Modeling

Principles of CAFE modeling are found in previously published articles [5-6, 14]. It is only briefly presented hereafter in the simplified case of a 2D static columnar solidification front growing in an undercooled liquid, which is pertinent for the present contribution.

The FE mesh is made of non structured triangles. The CA grid is made of regular square cells. Both triangles and cells remain fixed in space over time. The CA cells are much smaller than the finite elements. In a time stepping loop, the macroscopic conservation equations for total mass, energy, mass of solute and momentum are solved based on the finite element method. The enthalpy, composition and liquid velocity fields are interpolated from the FE nodes onto the CA cells. Due to the nucleation and growth algorithms, cells are captured. The size of the growing shapes associated to each cell defines a value for the volume fraction of the mushy zone within the cell, g^m. A microsegregation rule is then applied to each cell in order to deduce the temperature, T, and the internal volume fraction of solid, $g^{s\,m}$, considering the local average composition and enthalpy. The volume fraction of solid, g^s, is simply given by the product of volume fraction of the mushy zone and the internal volume fraction of the solid.

Implementation of the coupling scheme between the CA and FE models is validated according to the following procedure presented in Reference [8]. The growth undercooling of the columnar front is artificially decreased to almost zero by choosing adequate dummy values for a simple growth kinetics law. As a result, the growth front is developing very close to the local liquidus temperature. This situation is nothing but the one implicitly described by a purely macroscopic FE model. The results of the simulations carried out with the purely macroscopic FE model and the CAFE model are almost superimposed in time and space. The coupling scheme is thus validated and the model can be used with realistic parameters for the dendrite tip growth kinetics, accounting for the solute and curvature contribution to the undercooling as well as the intensity and orientation of the liquid velocity field with respect to the orientation of the crystal [13].

Results

Table I gives a detailed list of the values of the parameters and thermo-physical properties used for a CAFE simulation. The result of the predictions of the CAFE model is shown as a snapshot 165 minutes after the beginning of the simulation in Figure 2. The isotherms in Figure 2a are roughly vertical due to the one-dimensional configuration of heat extraction. The effect of counterclockwise convection in the liquid is yet slightly visible by the bending of isotherm 24°C. While the position of the mushy zone growth front displayed as a thick black line in Figure 2a-c agrees fairly well with observation in Figure 1, a pocket of liquid remains as shown by the black contour at the top-left corner of the figures. It corresponds to a zone of highly segregated liquid. Zones of dark grey level also appear in the mushy zone of Figure 1. In order to explain the formation of such a channel, the sequence of the formation of the mushy zone - liquid growth front is presented in Figure 3b. It is compared with the sequence recorded by X-Ray imaging shown in Figure 3a. As can be seen, the formation of such a channel is due to the destabilization of the growth front. It is highly dependent on the orientation of the crystal. Comparison with a simulation using a purely FE model demonstrates the need to account for the undercooling of the

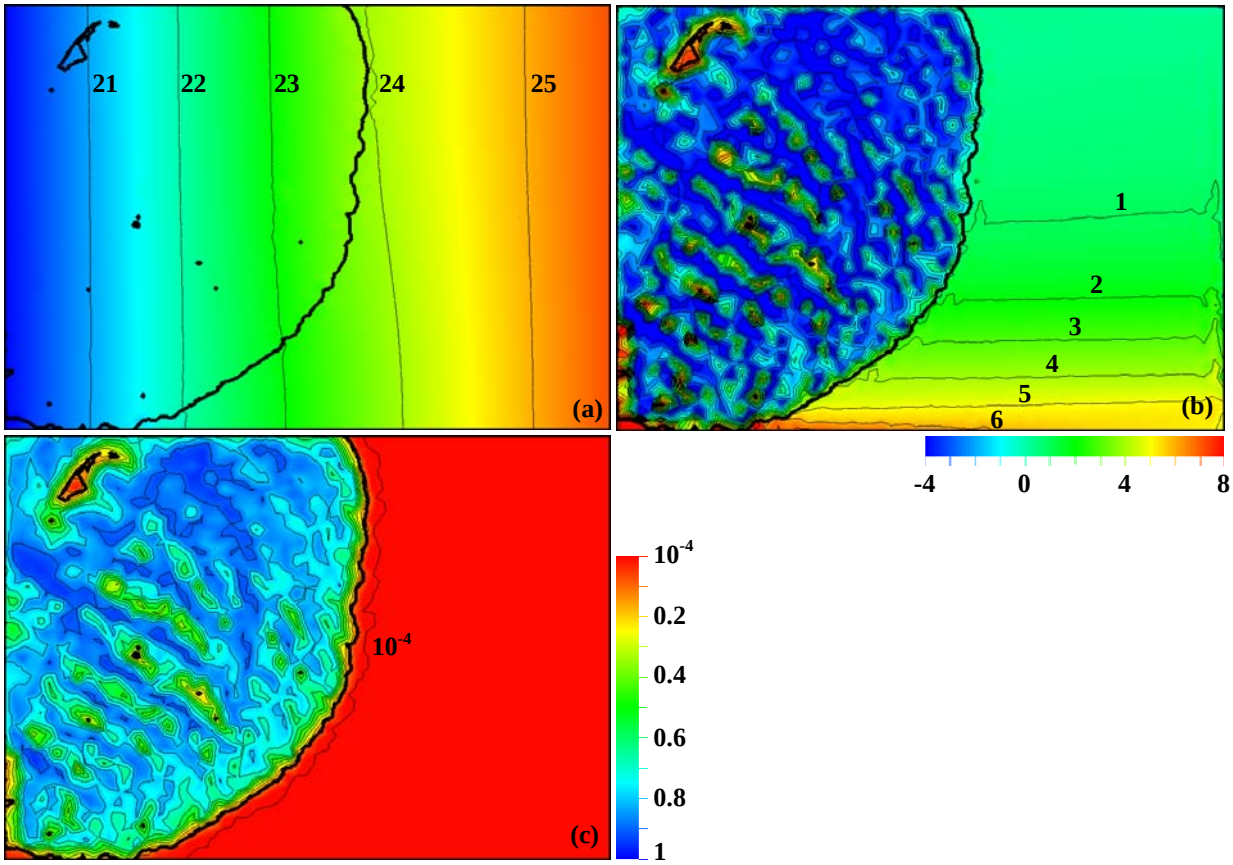

Figure 2. Predictions of the CAFE model for the solidification of a Gallium-5wt%Indium alloy showing maps of (a) temperature [°C], (b) relative composition, $\bar{w}$-w_0 [wt%] and (c) volume fraction of solid fraction, g^s [1], 165 minutes after the beginning of the simulation. The position of the growth front deduced from the cellular automaton model is superimposed on the figures as a thick black line. All values of data and parameters used for the simulation are listed in Table I.

growth front in order to simulate such phenomena. Indeed, Figure 3c shows a poor reproduction of the observed sequence. Since no nucleation undercooling is taken into account in the FE simulation, solidification starts as soon as the liquidus temperature of the Gallium-5wt%Indium alloy is reached. The front propagates rapidly in the FE simulation. The temperature gradient being limited, the casting is half mushy at time 110 minutes when the nucleation undercooling is reached. It is only at that time that nucleation takes place and solid starts to form with the CAFE simulation. Additional segregated channels are more clearly visible in Figure 2b, with traces of their formation with time shown as thick black lines in Figure 3b. These channels are oriented with respect to the modeled <10> directions of the crystal. As for the previous channel, they are a trace of the segregation between leading dendrites trunks forming at the growth front. These channels are to be compared to the distributions of the grey levels shown in the mushy zone of Figure 1. They are directly linked with patterns on the map of the fraction of solid in Figure 2c. Figure 3b clearly shows that the CAFE model overestimates the growth rate of the grain structure after nucleation between 110 and 150 minutes. Several reasons could be given to explain this drawback. They are mainly linked to the material properties that are largely unknown for the Gallium-Indium system. It includes the fact that Gallium solidifies with an orthorhombic structure. Its properties such as the diffusion coefficient of Indium in liquid Gallium was only estimated [15]. And no data was found for the Gibbs-Thomson coefficient, some value being used for this study. Also the model itself does only consider a constant value

for the dendrite arm spacing entering the permeability model of the mushy zone. The value is most probably too large at the onset of solidification when a large growth rate is recorded. As a consequence, the fluid flow is probably overestimated, also explaining the overestimation of the growth kinetics.

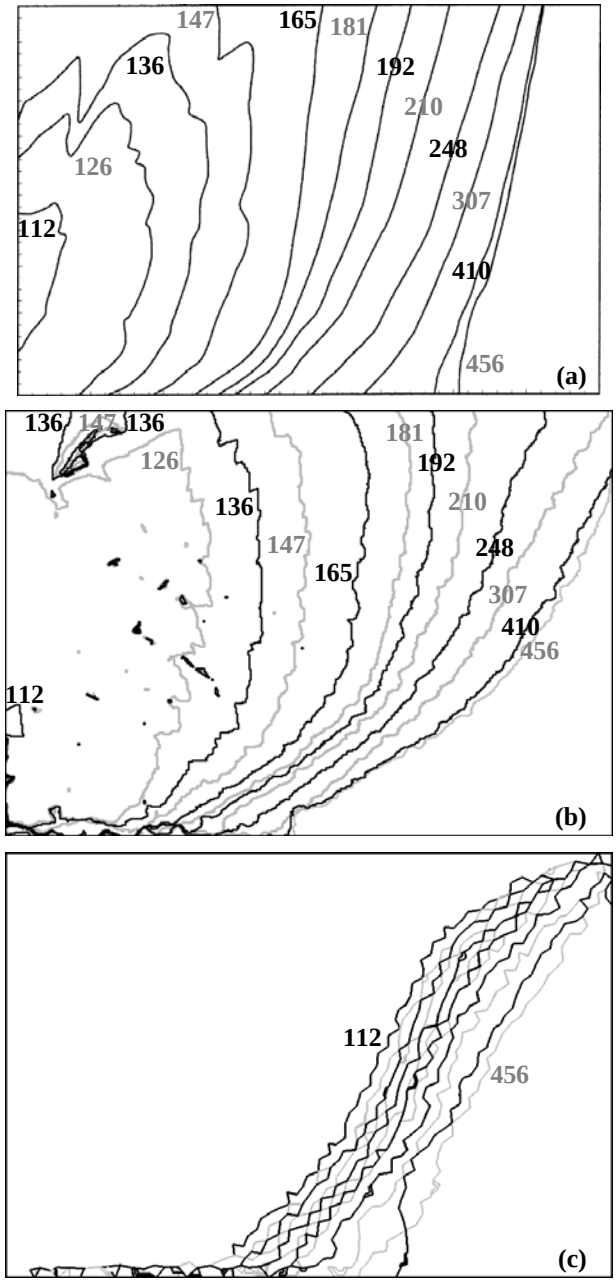

Figure 3. Sequences of the growth of the boundary between the mushy zone and the liquid domains during the solidification of the Gallium-5wt%Indium alloy (a) deduced from X-ray observations [2], (b) simulated using the CAFE model and (c) simulated using a FE model (growth front position is drawn with isofraction of solid 10^{-4}). Labels are times in minutes indicated from the beginning of the solidification experiment.

Stratification of Indium layers in the liquid domain observed in Figure 1 is also reproduced in Figure 2b. Calculated composition profiles along a vertical line of the liquid domain located 2 mm from the right-hand-side wall at time 165 minutes are drawn in Figure 4a for the FE and the CAFE simulations. They are compared with the data provided by Yin and Koster [2]. Due to

the higher density of Indium-rich liquid, Indium layers form at the bottom of the casting, which is well reproduced by the two simulations. The CAFE simulation predicts a sharper decrease of the Indium composition compared to the FE simulation. The In composition decreases to 6 wt% at the middle of the ingot height and then stabilizes to 5.5 wt% at the top. The FE simulation predicts a closer agreement to the experimental data. This is rather surprising when considering that the mushy zone at the time when the profile is drawn is predicted to fill the casting up to the top part of the ingot (Figure 3c). Indeed, a slope change of the profile at height 25 mm is due the domain change from liquid at the bottom to mushy one at the top. Such a slope change is not observed in the measured profile. This finding could be explained either by an overestimation of the solute content in the mushy zone by the CAFE model and/or by the calibration procedure used by Yin and Koster [2] to convert the signal coming from X-Ray imaging into a composition map. Comparison of the cooling curves is also available in Figure 4b. Simulations results for the FE and CAFE simulations are superimposed at locations A and C since Dirichlet boundary conditions were imposed using the average cooling rates measured there. At a time close to 165 minutes, thermocouple B experiences a decrease of the cooling rate. This is also reproduced by the CAFE simulation, with some overestimation. As shown in Figures 1 and 2b, the growth front passes position B at about the same time. The decrease of the cooling rate is thus nothing but the consequence of the large latent heat release associated with the development of the growth front. The FE simulation also predicts the effect of the latent heat release but at about 50 minutes. This is due to the immediate development of the mushy zone with no consideration of the nucleation undercooling that delays solidification.

Summary

This contribution is a first comparison of predictions using a CAFE model with in-situ observation of the development of both segregation and structure forming during a solidification experiment. It clearly reveals the limitation of using a purely macroscopic FE model to predict the dynamics of the mushy zone growth front based on the position of the isofractions of solid. Destabilization of the growth front is shown to take place, which are the origin of segregated channels observed in the solidifying mushy zone. These results confirm the conclusions given in a previous contribution that measurement of macrosegregation maps should be carried out at the scale of the grain structure if one whishes to study the interaction between the formation of the structure and the segregation at the scale of the casting [8]. The present set of data is unique for the validation of the predictions of the CAFE. However, no information is given on the final segregation map of the ingot in its as-cast state. Such post-mortem measurements are required for further comparison. It is also suggested that new investigations should be carried out using cubic materials as suggested in Reference [18, 19]. This would permit more links with most of the industrial alloys cast concerning both segregation path and crystallographic structures. Finally, modeling efforts should be carried out to develop a 3D version of the CAFE coupling scheme for improved predictions of the link between structure and segregation formed in casting.

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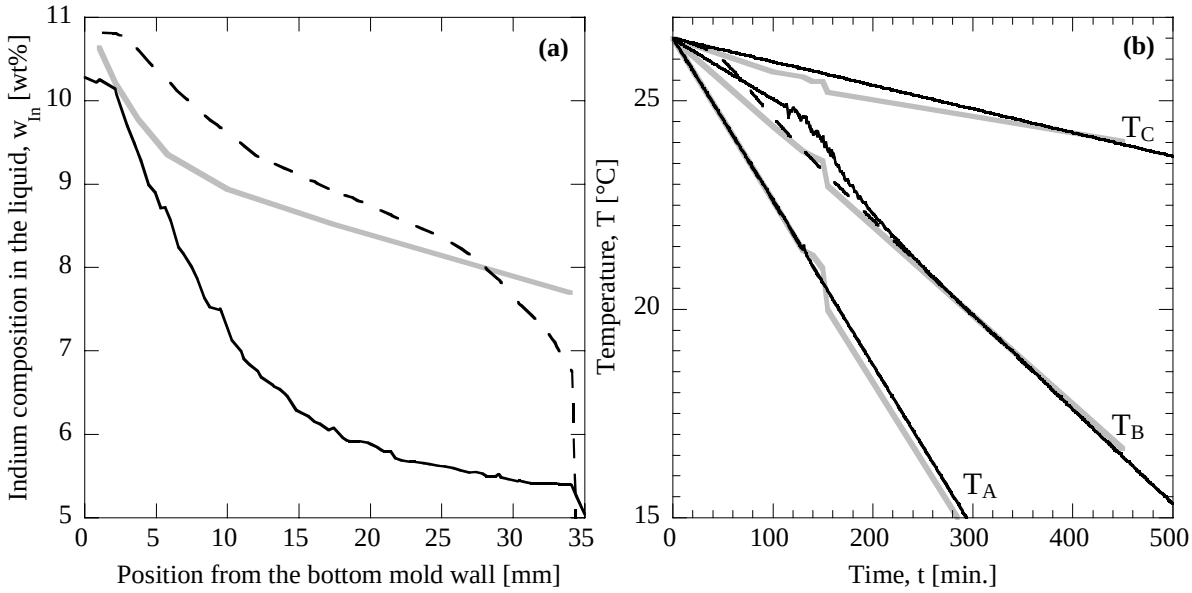

Figure 4. Comparison of measured versus predicted (a) composition profile along a vertical line in the remaining liquid at time 165 minutes [2] and (b) temperature history at locations of thermocouples A, B, and C shown in Figure 1 [1]. Curve style: (grey) measured, (plain) simulated using the CAFE model, (dashed) simulated using a FE model.

Table I: Value of thermo-physical data and numerical parameters

Latent heat of fusion [16]	J kg^{-1}	$8.024\ 10^4$	Initial temperature [1]*	°C	26.5
Heat capacity [15]*	J kg^{-1} °C^{-1}	380.74	Left-hand-side cooling rate [1]*	°C h^{-1}	-2.362
Thermal conductivity [15]	W m^{-1} °C^{-1}	29.09	Right-hand-side cooling rate [1]*	°C h^{-1}	-0.328
Melting temperature [2]	°C	29.8	Number of FE nodes	-	2442
Eutectic temperature [2]	°C	15.3	Time step (macro)	s	0.1
Liquidus slope [2]*	°C wt%$^{-1}$	-0.677	***Additional parameters for a CAFE simulation***		
Segregation coefficient [2]	-	0	Diffusion coeff. (In in liq. Ga) [15]	m^2 s^{-1}	$1.525\ 10^{-9}$
Eutectic composition [2]*	wt%	21.42	Gibbs-Thomson coefficient	K m	$4.34\ 10^{-5}$
Liquid density [1]	kg m^{-3}	6129.7	Nucleation undercooling [1]*	°C	4.25
Nominal In composition [1]	wt%	5	Position of nucleus [1]×*	m; m	0; $9\ 10^{-3}$
Thermal expansion coefficient [15]	°C^{-1}	$1.19\ 10^{-4}$	Grain orientation [1]+*	°	- 30
Solute expansion coefficient [15]*	wt%$^{-1}$	$-1.663\ 10^{-3}$	Time of nucleation event [1]	s	110
Dynamic viscosity [15]*	Pa s	$2.03\ 10^{-3}$	Cell sizew	m	10^{-4}
Dendrite arm spacing	m	500			
* deduced from			× horizontal and vertical distance from the bottom-left-corner in Figure 1		
+ with respect to the horizontal axis					

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